

REMARKS

Claims 1, 4, 6-10 and 12 are pending. Claims 2, 3, 5, 11 and 13 have been canceled.

Claims 1 and 12 have been amended. The phrase "while inhibiting by-production of acetaldehyde by increasing carbon monoxide partial pressure or methyl acetate content," has been added to better define the invention. Also, the wording "at a water content of the reaction mixture of 3 percent by weight or more" has been changed to "at a water content of the reaction mixture of 3 percent by weight or less". The word "less" is the correct expression in this section. Support for this amendment appears in the original claims and on page 15, lines 10-12.

Claim 4 has been amended by replacing the wording "3.1 percent by weight or more" with "3.1 to 10 percent by weight" so that claim 4 further limits claim 1.

No new matter has been added by way of the above-amendment.

Prior Art Based Issues

Claims 1, 2, 4, 6-10, and 12 are rejected under 35 U.S.C. 103(a) as being unpatentable over Hosono et al. (US 2006/0281944), Scates et al. (US 6,303,813) or Jones (US 7,098,363) all in combination, and further in view of Miura et al. (EP 0687662) and Cheung et al. (US 7,005,541). Applicants respectfully traverse the rejection.

(1) Present Invention

Claim 1, as currently amended, recites as follows:

A method for producing acetic acid while inhibiting by-production of acetaldehyde by increasing the carbon monoxide partial pressure or the methyl acetate content, comprising the steps of:

continuously reacting methanol with carbon monoxide in the presence of a homogeneous rhodium catalyst, an iodide salt, methyl iodide, methyl acetate, and water;

producing acetic acid at a production rate of 11 mol/L·hr or more;

keeping the acetaldehyde content of a reaction mixture to 500 ppm or less; and

keeping the production rate of acetaldehyde to 1/1500 or less of the production rate of acetic acid,

wherein the reaction is carried out at a carbon monoxide partial pressure in a gaseous phase of a reactor of 1.05 MPa or more and at a methyl acetate content of the reaction mixture of 2 to 10 percent by weight and at a water content of the reaction mixture of 3 percent by weight or less and at a hydrogen partial pressure in the gaseous phase of the reactor of 70 kPa or less.¹

Applicants respectfully submit that this combination of features is neither taught nor fairly suggested by the cited prior art.

The Examiner asserts that it is easy to combine the components disclosed in the cited references, namely, a homogeneous rhodium catalyst, methyl iodide, methyl acetate, methanol, carbon monoxide and the like to complete the present invention. However, these constituents do

¹ Claim 12 is similar to claim 1, but claim 12 defines the rhodium material in a different way than claim 1.

not simply have an additive effect (in other words, the effects do not simply pile up due to their existence), e.g., the effect of a certain element could be to increase the production of the main products while also increasing the production of the by-products, and as a result, various combinations result in various and complex effects. It is Applicants' position that the artisan would have insufficient guidance based on the cited references to obtain the inventive method, in view of the fact that an excessive amount of experimentation would be necessary to achieve an optimal condition with various combinations of experimental conditions. Here, the Examiner fails to consider these competing factors which would weigh heavily on the artisan.

One of the features of the present invention is "to provide a method for efficiently producing high-quality acetic acid, which method can reduce production of by-products without reducing the reaction rate of acetic acid" (see page 8, line 21-24 of the specification) on the basis of the fact that "only the CO partial pressure and the methyl acetate content serve to accelerate the main reaction while inhibiting side reactions" (page 13, line 17-19 of the specification). In order to clarify the above feature, Applicants have amended claims 1 and 12 to recite the wording "while inhibiting by-production of acetaldehyde by increasing the carbon monoxide partial pressure or the methyl acetate content".

In the methods for producing acetic acid, it may be preferable to keep the concentration of acetaldehyde as a by-product to a low level, and some of the prior art references may disclose the preference of such a low concentration of acetaldehyde. However, even if the prior art references disclose that the lower concentrations of acetaldehyde is preferred (for instance, 400 ppm or less), merely combining the description of the reference and the common technology in the art would not lead to the completion of the present invention. Furthermore, it is unexpected that the present invention has such incredible effects.

Factors involved in the reaction for producing acetic acid include reaction temperature, CO partial pressure, hydrogen partial pressure, methyl iodide content, methyl acetate content, water content, rhodium content and lithium iodide content. All of them naturally influence the concentration of acetic acid -- the main reaction product. However, what kind of effects these factors have in producing the main by-product were unknown to anyone prior to Applicants' disclosure. The present inventors have performed the extensive experiments for the first time,

and found that only the CO partial pressure and the methyl acetate content serve to accelerate the main reaction while inhibiting side reactions.

(2) Cited References

In the outstanding Office Action, the Examiner cited five references, Hosono et al., Scates et al., Jones, Miura et al. and Cheung et al.

Hosono et al. teach that acetic acid productivity decreases when the carbon monoxide partial pressure is below 1.0 MPa. However, the reference is silent with regard to the relationship among CO partial pressure, methyl acetate content and concentration of the methyl acetate. In addition, the reference is silent with regard to the mutual relationship among other components in the reactant mixture, namely, reaction temperature, hydrogen partial pressure, methyl iodide content, water content, rhodium content and lithium iodide content.

Scates et al. teach a method for producing acetic acid by a carbonylation process using a homogeneous rhodium catalyst with methyl iodide, and describes that the formation of by-products such as aldehydes is decreased by keeping the hydrogen partial pressure of about 0.1 to 4 psia. However, upon reviewing Table II of Scates et al., it is noted that the increase in the reaction temperature leads to the keeping of the acetic acid production rate (acetic acid production rate corresponds to methanol feed (g/min), and methanol feed (g/min) is proportional to acetic acid production rate provided that methyl acetate concentration is constant). Also, generally, when hydrogen partial pressure decreases, acetic acid production rate increases (please see Table 1 of the present application). In Scates et al., methanol feed is decreased and methyl acetate concentration is increased (Example 1) because the produced acetic acid reacted with methanol to result in methyl acetate. Indeed, Scates et al. states that the hydrogen partial pressure of 4 psia or less can decrease the formation of by-products such as aldehydes. However, Scates et al. is quite different from the present invention which enhances the acetic acid production rate while inhibiting the by-production of acetaldehyde in that the former accompanies the decrease in the acetic acid production rate. Therefore, Scates et al. requires at the same time another means (for example, increase in reaction temperature) for industrial use.

Jones teaches a faster reaction rate for carboxylic acid production by adjusting methyl acetate concentrations. However, Jones fails to describe the content of acetaldehyde in the reaction mixture.

Miura et al. teach a process for producing high purity acetic acid by carbonylation of methanol in the presence of a rhodium catalyst, an iodide salt, and methyl iodide. However, the reference is silent with regard to the relationship among CO partial pressure, methyl acetate content and concentration of acetaldehyde.

Cheung et al. teach further purification steps by way of plural distillation, and refers to the relationships among water, rhodium catalyst and methyl acetate. However, the reference is silent with regard to the relationship between CO partial pressure and concentration of acetaldehyde.

Thus, the references cited by the Examiner each disclose individual parts of the inventive production conditions. However, these fragments of information cannot be assembled to complete the present invention, since the present invention is based on the special technology of "enhancing only the CO partial pressure and the methyl acetate content to accelerate the main reaction while inhibiting side reactions" in the method for producing acetic acid according to the present invention, wherein the method comprises the steps of: continuously reacting methanol with carbon monoxide in the presence of a homogeneous rhodium catalyst, an iodide salt, methyl iodide, methyl acetate, and water; and keeping the acetaldehyde content of a reaction mixture to 500 ppm or less. In order to combine the experimental knowledge to complete the present invention from the combination of the descriptions of the cited references, it is absolutely necessary to have a technical idea of "enhancing only the CO partial pressure and the methyl acetate content to accelerate the main reaction while inhibiting side reactions". However, the cited references along with other prior-art methodologies decisively lack such a teaching or suggestion.

Based only on the fragmentary knowledge of "lower concentration of by-product acetaldehyde" and "that acetic acid productivity decreases when the carbon monoxide partial pressure is below 1.0 MPa", the Examiner asserts that the prior-art knowledge can be easily combined and those skilled in the art can easily complete the present invention. Applicants

respectfully submit that the Examiner is using improper hindsight reconstruction. The text of section 103, includes the phrase "at the time the invention was made." For it is this phrase that guards against entry into the "tempting but forbidden zone of hindsight," see *Loctite Corporation v. Ultraseal Ltd., et al.*, 228 USPQ 90 (Fed. Cir. 1985), when analyzing the patentability of claims pursuant to that section. Measuring a claimed invention against the standard established by section 103 requires the oft-difficult but critical step of casting the mind back to the time of invention, to consider the thinking of one of ordinary skill in the art, guided only by the prior art references and the then-accepted wisdom in the field, see, e.g., *W.L. Gore & Associates, Inc. v. Garlock, Inc.*, 220 USPQ 303 (Fed. Cir. 1983). The case law makes clear that the best defense against the subtle but powerful attraction of a hindsight-based obviousness analysis is rigorous application of the requirement for a showing of the teaching or motivation to make the modifications to a reference's teachings necessary to obtain the present invention, see for e.g., *In re Fine*, 5 USPQ2d 1596 (Fed. Cir. 1988).

Here, at the time of the present invention, the combination of the elements of the present method was not fairly suggested by the cited prior art, and such a combination was found out through an extensive, diligent effort on the part of the inventors.

Furthermore, there would be no expectation of success by the artisan in combining the various elements from the cited references, in view of the undue amount of experimentation which would have been necessary to complete the present invention. For the sake of gathering and selecting the limited number of suitable parameters from a myriad of parameters, the artisan would need some motivation or reason for selecting the specific set of the parameters to obtain the present invention.

As part of the invention, the present claims require "*a carbon monoxide partial pressure in a gaseous phase of a reactor mixture of 1.05 MPa or more, a methyl acetate content of the reaction mixture of 2 to 10 percent by weight, a production rate of 11 mol/L · hr or more and acetaldehyde content of 500 ppm or less*". Obtaining this combination of elements was the result of extensive experiments, and was not the result of optimization of selected conditions, as asserted by the Examiner. It is impossible to combine these numerical values at once by chance even if there is a document that describes these numerical values. Because there are such a

myriad of parameters, the combination of these parameters runs into astronomical numbers. If the Examiner asserts that the present invention is obvious, the Examiner is requested to clarify the reason why one skilled in the art would select the claimed conditions of "*a carbon monoxide partial pressure in a gaseous phase of a reactor mixture of 1.05 MPa or more, a methyl acetate content of the reaction mixture of 2 to 10 percent by weight, a production rate of 11 mol/L · hr or more and acetaldehyde content of 500 ppm or less*". If the Examiner is not able to further clarify the reason an artisan would select the present invention, then the rejection would be untenable and based on improper hindsight reconstruction.

Here, the Examiner has identified five references which the Examiner alleges contains all of the elements of the instant claims. However, simply showing that all of the elements of the application/patent claim are present in the prior art does not necessarily mean that the invention is obvious. "This is so because inventions in most, if not all, instances rely upon building blocks long since uncovered, and claimed discoveries almost of necessity will be combinations of what, in some sense, is already known." *KSR International Co. v. Teleflex Inc.*, 82 USPQ2d 1385, 1389 (U.S. 2007).

In *KSR Int'l*, the Supreme Court emphasized that a reason for the skilled artisan to do what is claimed is part of the obviousness inquiry. Justice Kennedy states: "As is clear from cases such as *Adams* [383 U.S. 39, (1966)], a patent composed of several elements is not proved obvious merely by demonstrating that each of its elements was, independently, known in the prior art...it can be important to identify a **reason** [or rationale] that would have prompted a person of ordinary skill in the relevant field to combine the elements in the way the claimed new invention does." *KSR International Co.* at 1396. (Emphasis added).

In *KSR Int'l*, the Supreme Court gave several rationales for use in the obviousness analysis (see rationales A-G in the "Guidelines" in MPEP 2141). In the present case, the Examiner's rationale is similar to Rationale (A) which is the combining of prior art elements according to known methods to yield predictable results. However, this rationale is not proper in arts that are relatively unpredictable. The obviousness standard based on the *KSR v. Teleflex* case involved a mechanical device in a relatively predictable technological area. The courts recognize that inventors face additional barriers in relatively unpredictable technological areas such as the

chemical area as noted in *Takeda Chemical Industries, Ltd. v. Alphapharm Pty., Ltd.*, 83 USPQ2d 1169 (Fed. Cir. 2007).

Here, the present method is nonobvious, since the artisan would have no expectation of success to pick and choose from the various parameters from different references in an unpredictable technology.

Additionally, even though there is no *per se* rule of a maximum number of references which can be combined in an obviousness rejection, when documents to be combined are not just two but five, like in the present case, that should be a signal that the present invention is leaning towards nonobviousness. It is usually more difficult to combine the knowledge and obtain suitable effects of a specific combination when the number of documents which are to be brought together increases. This is due to the fact that increasing the number of documents results in necessarily results in an increase in the number of parameters. Here, one must review an astronomical number of the combinations of parameters from the cited references, since each reference discloses a range of plural parameters. In this sense, the Examiner's assertion is in error, and the present invention is nonobvious.

(3) Homogeneous catalyst

Applicants respectfully submit that the cited references do not combine to render the present method obvious, since some of the references are drawn to catalysis using a homogeneous catalyst whereas some of the references (Hosono et al. and Jones) are drawn to catalysis using a heterogeneous catalyst in the reaction.

The present invention is characterized in the use of the homogeneous catalyst.

One skilled in the art would not use a homogeneous catalyst in place of the heterogeneous catalyst, or vice versa. In addition, one skilled in the art would not use the reaction conditions associated with a process involving a homogeneous catalyst in place of the reaction conditions associated with a process involving a heterogeneous catalyst, or vice versa. Because the reaction conditions and the reaction mechanism are greatly different between the homogeneous catalyst and the heterogeneous catalyst, the conditions described for use with the

heterogeneous catalyst cannot easily be replaced with the conditions described for the homogeneous catalyst. For instance, the differences between the homogeneous catalyst and the heterogeneous catalyst is described in "Introduction to homogeneous catalyst and heterogeneous catalyst"² (in Japanese) (attached material 1). Below is a partial translation of the relevant parts:

- A transition metal complex which is soluble in the solvent is used as a homogeneous catalyst. Reactions using the homogeneous catalyst is performed in liquid phase, the reaction temperature is usually 200°C or less. The catalytic activity of the homogeneous catalyst is lower than that of the heterogeneous catalyst but the selectivity of the reaction with homogeneous catalyst is higher than that of the heterogeneous catalyst (page 4, lines 6-8).
- Usual heterogeneous catalysis reaction is performed by contacting a gassy reactant with a solid catalyst, the reaction temperature relatively high (250-550°C), and catalytic activity is also high (page 4, lines 4-5).
- In case of homogeneous catalysis, the catalytically active species is generally a single metal complex (page 4, lines 16).
- In case of heterogeneous reaction, there is a possibility that many types of the active site exists on the surface (page 5, lines 1-2).

From the above-mentioned viewpoint, it is clear that the results accomplished by the present invention using the homogeneous catalyst would not be expected from the teachings of the cited references alone, and that the present invention that uses the homogeneous solvent cannot be easily completed from the teachings of the cited references alone. Moreover, it would be necessary to perform undue experiments to complete the present invention. Therefore, the present invention that uses the homogeneous catalyst/solvent is nonobvious over the cited references. Reconsideration and withdrawal of the rejection are respectfully requested.

In view of the above amendment, Applicants believe the pending application is in condition for allowance.

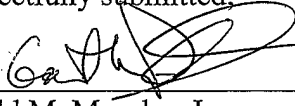
² Hitai, Masanobu and Ichikawa, Masaru. Introduction to homogeneous catalyst and heterogeneous catalyst. Maruzen Co. Ltd., 1983, p. 4-6.

Should there be any outstanding matters that need to be resolved in the present application, the Examiner is respectfully requested to contact Garth M. Dahlen, Ph.D., Esq., Reg. No. 43,575, at the telephone number of the undersigned below, to conduct an interview in an effort to expedite prosecution in connection with the present application.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37.C.F.R. §§1.16 or 1.17; particularly, extension of time fees.

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Attached: Hitai, et al. Introduction to homogeneous catalyst and heterogeneous catalyst. Maruzen Co. Ltd., 1983, p. 4-6.

化学セミナー11

均一触媒と不均一触媒入門

——これからの触媒化学——

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1・2 均一触媒と不均一触媒

触媒はその作用状態により均一および不均一触媒に分類される。一般に、金属化合物あるいは無機酸化物に担持された金属が不均一触媒として用いられる。ふつうの不均一反応はガス状にした反応基質を固体触媒と接触させ行わせる。反応温度は比較的高く (250~550°C)、触媒活性も大きい (表 1・1)。

表 1・1 均一および不均一触媒の比較

| 比較項目 | 均 一 | 不均 一 |
|-----------|-------------------|-------------------------|
| 触媒の形 | 可溶な(単核の)遷移金属錯体 | 無機酸化物に担持された金属あるいは金属酸化物 |
| 触媒の(触)安定性 | 低い | 高い |
| 反応相 | 液相 | 気相/固相 |
| 反応温度 | 低い(200°C以下) | 高い(250~550°C) |
| 触媒の活性 | 低い | 高い |
| 反応の選択率 | 高い | 低い |
| 触媒の分離 | 分子レベルでかなり明確に理解される | 分子レベルでの反応機構の解明はかなり困難である |

一方、均一触媒としては溶液中に可溶な遷移金属錯体を用いられる。反応は均一な液相で行われ、反応温度はふつう 200°C 以下である。触媒活性は不均一触媒に比べて低いが、選択率が高いのが特徴である。均一触媒の不利な点は、空気、湿度、温度などに不安定な高価な触媒を反応終了後に生成物と分離しなければならぬところにある。

水素化、異性化、カルボニル化、不均化、酸および重合反応など多くの反応は均一および不均一触媒の両方で進行する。しかし、飽和炭化水素の C—C および C—H 結合の活性化や CO および N₂ の三重結合の水素化については、まだ効率のよい均一触媒は知られていない。

反応機構に関しては、均一触媒を用いた反応のほうがより分子レベルで理解される。すなわち、均一触媒反応の場合は一般に反応活性種は単一の金属錯体であり、赤外あるいは NMR などの物理化学的手段により反応を追跡することもできるからである。また、ある場合には反応中間体を単離し、X 線構造解析

などを行うことも可能である。一方、不均一触媒反応の場合はいくつもの型の活性点が表面に存在している可能性がある。最近では AES (Auger electron spectroscopy), UPS (ultraviolet photoelectron spectroscopy), LEIS (low energy electron diffraction) など多くの固体表面分析に有効な機器が開発され、反応機構がかなり詳細に議論できるようになった。また、均一触媒反応における研究成果が不均一触媒反応の機構解明に大きく貢献している点も見逃せない。

エチレンの水素化を例に取り上げ、均一および不均一触媒を用いた場合の反応機構を比較してみよう (図 1・4)。どちらの反応も M—H 結合へのエチレンのそう入による M—C₂H₅ 基の生成とそれにつづく M—H との遷移的脱離に

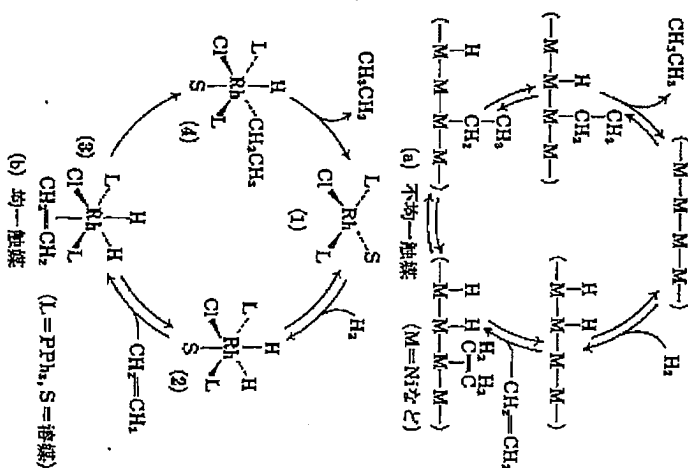


図 1・4 エチレンの水素化

よるエタンの生成から説明される。しかし、分子レベルで見た場合に、均一触媒のほうがより明確に理解されていることがわかるであろう。

2 遷移金属錯体の基本的反応

遷移金属は、原子価殻に一部分でんしたd軌道あるいはf軌道をもつため、反応基質と様々な形式で結合することができ、また、遷移金属の配位圏内で行われる独特の反応を起こし、これらが均一触媒反応の重要な素反応となる[†]。以下、遷移金属錯体に特有のいくつかの基本的反応を解説しよう。

2・1 金属への配位と解離

遷移金属はいろいろな分子あるいは化合物(S)をその配位圏に取り込み、遊離の場合と異なる状態にすることができる。また、この逆のSが金属の配位圏からはなれる反応は解離とよばれる[式(2・1)]。



[†] このあたりの基礎知識は、F. R. Hartley, "Elements of Organometallic Chemistry", The Chemical Society (1974)；千鶴貞信 訳, "化学セミナー7 有機金属化学の基礎", 丸善 (1981) を参照されたい。

| | | | |
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化学セミナー 11
 均一触媒と不均一触媒入門
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